

MITIGATION OF HEAVY METALS IN VEGETABLES THROUGH WASHING WITH HOUSE HOLD CHEMICALS

**MUHAMMAD UMAIR SATTAR, MUHAMMAD ASIF KHAN, ANEES AHMED KHALIL &
RAI MUHAMMAD AMIR**

National Institute of Food Science and Technology, University of Agriculture, Faisalabad, Pakistan

ABSTRACT

Vegetables play key role in maintaining the nutritional status of humans. The dominant elements in vegetables are calcium, potassium, iron and sodium etc. which provide alkalizing effects, neutralizing the acidity produced by other foods, especially those of animal origin. Heavy metals in vegetables accumulate through uptake from soil, water and atmosphere. The intake of heavy metals through diet leads to several disorders such as kidney damage, nervous disorder, bone disease and tubular growth. In Pakistan this situation is more alarming as vegetables grown in peri-urban areas have shown high incidence of heavy metals accumulation. In this study effort was made to mitigate different heavy metals (Cd, Cr, As, Pb and Hg) in cauliflower, spinach, okra and brinjal collected from self-grown, wastewater irrigated, supervised field through chemical washing techniques. Heavy metals contents were determined by using Atomic Absorption Spectrophotometry (AAS). The data was subjected to statistical analysis to identify the most efficient washing treatment for heavy metal mitigation. Vegetable showed high load of heavy metals in unwashed form that reduced significantly by different washing techniques. Among the chemical washing techniques, acetic acid 10% was found to be more effective.

KEYWORDS: Heavy Metals, Vegetable, Chemical Washing, Wastewater

INTRODUCTION

Food safety is a major public concern worldwide. This is serious problem in the whole world especially in economically poor countries. The production of safe food is an important aspect of food quality assurance as well as human health. The people are demanding fresh, hygienic and quality food. During the last few decades, the increasing awareness of food safety has stimulated to do research on the risk associated with consumption of foodstuffs contaminated by pesticides, heavy metals residues and toxins (D'Mello, 2003). Agriculture is backbone of Pakistan's economy and contributes greatly towards GDP in Pakistan. Up to 70 % of our rural population is directly or indirectly engaged in this sector. In Pakistan only 2 percent of the total cultivated area is under vegetable cultivation and it contributes about 0.22% in total world vegetable overall export. The per capita daily intake of vegetables is 134g, which is 33-35% below the minimum recommended level of 200g per day (GOP, 2011). The vegetables are a valuable source of minerals, vitamins, fibers and contribute significantly to human health. Vegetables also have some functional constituents such as protein, vitamins and minerals which have health promoting role. Consumer's demand for good quality and safe vegetables is increasing (Sobukola and Dairo, 2007).

Vegetables in Human Nutrition

Vegetables are important in human diet because of their valuable contribution in terms of colors, fragrance and taste in food. The vegetables have also beneficial role in growth and development of the body. These not only strengthen the immune system of the body but also help in disease prevention. The edible portion of vegetable includes stem, leave,

fruit, seed and roots that mostly require cooking for maximum digestion (Wargovich, 2000). Cauliflower, spinach, okra and brinjal are most common vegetables consumed by the people in Pakistan. Cauliflower possess a high nutritional profile as it is rich in water, vitamins, folate and dietary fibers but less in fat. Green leafy vegetables contribute sufficient quantity of vitamins and minerals to human diet. They are good source of iron, calcium, phosphorus, folic acid, riboflavin, ascorbic acid and carotene. The vegetables contain high water content, sugars, protein, starch, fat and energy value (Munteanu *et al.*, 2011). Vegetables are considered as natural reserves of nutrients gifted by Almighty Allah to human beings e.g. carrot is a valuable source of vitamin A, needed for normal vision, like wise spinach and tomato contains sufficient amount of vitamin C to prevent and cure scurvy. Potato is good in starches and provide high amount of carbohydrates. Some vegetables contain high amount of dietary fibers that helps to prevent constipation (Sonni Alvarez, 2002). The USDA 2000 Dietary Guidelines (USDA, 2000) recommended the consumers to: (1) Eat at least 3 servings of vegetables and 2 servings of fruits every day (2) choose fresh, frozen, dried, or canned vegetables (3) Incorporate dark-green leafy vegetables, orange fruits and vegetables and cooked dry beans and peas often. Vegetables also have phytochemicals that function as antioxidant, detoxifying agent, prevent tumor growth and helps to modify metabolic processes. Vegetables are economical way to deal with diseases those results from micronutrient deficiency such as kidney stones, skin diseases, night blindness, anemia, diseases of heart, lungs and breast (Wargovich, 2000).

Table 1: Nutritive Constituents of Vegetables

Constituent	Sources	Established or Proposed Effects on Human-Wellness
Vitamin C (ascorbic acid)	Broccoli, cabbage, cantaloupe, leafy greens, pepper, potato and tomato	Prevents scurvy, aids wound healing, healthy immune- system, cardiovascular-disease
Vitamin A (carotenoids)	Dark-green vegetables (such as spinach and turnip), vegetables (such as carrots and sweet potato) and tomato	Night blindness prevention, chronic fatigue, psoriasis, heart disease, stroke, cataracts
Vitamin K	Green onions, crucifers (cabbage, broccoli, brussel sprouts), leafy greens	Synthesis of pro-coagulant factors, osteoporosis
Vitamin E (tocopherols)	Chickpeas, dark-green leafy vegetables	Heart-disease, LDL-oxidation, immune-system, diabetes, cancer
Fiber	Fresh vegetables, peas and dark green leafy vegetables	Diabetes, heart disease
Folate (folicin or folic acid)	Dark-green leafy vegetables such as spinach, mustard greens, lettuce and broccoli	Birth defects, cancer heart disease, nervous system
Calcium	Cooked vegetables (such as beans, greens, okra, tomato, peas and cauliflower	Osteoporosis, muscular/ skeletal, teeth, blood pressure
Magnesium	Spinach, okra and potato	Osteoporosis, nervous system and immune system
Potassium	Baked potato or sweet potato	Hypertension (blood pressure) stroke and arteriosclerosis

Heavy Metals Accumulation in Vegetables

The heavy metals and their compounds rapidly contaminate agricultural land and becoming an alarming issue throughout the world. In different countries such as Australia and New Zealand, regulation and guidelines have been developed regarding the heavy metals that contaminate food chain and environment (McLaughlin *et al.*, 2000). It has been estimated that food of plant origin contributes about half of the mean ingestion of lead, cadmium and mercury (Itanna, 2002). Vegetables are also contaminated with heavy metals during transportation and marketing due to polluted air (Agrawal, 2003). Different agricultural practices such as use of fertilizer, manure etc. to enhance the crop yield are also heavy metals contributor in the soil. Soil pollution with toxic metals is becoming an alarming issue in Pakistan as it

disturbs the environmental safety (Bhutto *et al.*, 2009). There are number of factors that affect the metals uptake by the plant from the soil such as heavy metal concentration, type of the fertilizer applied, plant growth stage and species (Sharma *et al.*, 2006; Ismail *et al.*, 2011). Morpho-physiological nature of the vegetables greatly influence the quantity of the metal deposited on the plant surface (Singh and Kumar, 2006). Heavy metals are the most dangerous contaminants of soil ecosystem as well as food chain all over the world (Zahir *et al.*, 2009).

Sources of Heavy Metals

The sources of heavy metal contamination are waste water irrigation, the application of metal-based pesticides and fertilizers, industrial emissions and transportation. Wastewater irrigation is the major contributor of heavy metals contents of the soil (Mapanda *et al.*, 2005). High concentrations of heavy metals were reported in vegetables from the untreated wastewater irrigated areas (Sinha *et al.*, 2005; Sharma *et al.*, 2006). Use of industrial waste water for raising vegetables is very serious issue in Pakistan because these effluents are heavily loaded with harmful metals and metallic compounds (Singh *et al.*, 2004). Heavy metal pollution of agricultural land and crops in the surrounding area of mining has been considered as a primary environmental concern (Kalali *et al.*, 2011). Vegetable contamination with heavy metals is through absorption (from soil) and surface deposition (from polluted air). The plant species possess different potential to remove and accumulate different metals and results in serious health complications when such food stuff is consumed (Zurera *et al.*, 1989).

Health Implications of Heavy Metals

The intake of heavy metals for longer period of time not only results in the disruption of various biochemical activities but also adversely affect our vital body organs such as kidney, liver etc. In general humans are exposed to these metals by ingestion (drinking or eating) or inhalation (breathing). The metals may also be ingested involuntarily through food and drink. Dietary sources of heavy metals include contaminated food stuff such as fruits, vegetables and drinking water (Kachenko and Singh, 2006). The excessive intake of toxic metals can cause severe complications in humans and animals. Consumption of fruits and vegetables loaded with heavy metals such as Cd, Pb or even Cu and Zn are reported to cause cancer (Turkdogan *et al.*, 2002) and cancer of the pancreas, urinary bladder or prostate (Waalkes and Rehm, 1994). Lead, cadmium and chromium were found to be accumulated in the shoot and roots of plants at low, medium or high levels (Verma and Dubey, 2003; Yang *et al.*, 2003; Chandra and Kulshrestha, 2004; Adeyeye, 2005).

Arsenic, cadmium, chromium, mercury and lead are most prevalent metals that can pose threat to humans even at low concentration (Gupta, 1998). The arsenic is a toxic element and the humans may get exposure to arsenic through drinking water obtained by wells bored into arsenic contaminated areas or through contaminated water by agro-chemical waste or industrial effluents. Initial medical signs of severe arsenic poisoning include softness with tinting skin, muscular pain, vomiting with severe nausea, pain in stomach, and rice-water stools with diarrhea (Hughes *et al.*, 1988). Lead is considered as poison for humans who find its way into the human body through infected food stuff, air and water. Removal of lead by household washing practices of vegetables is also very rare (Muchuweti *et al.*, 2006). Cadmium is not required by the human body and due to its high specific gravity, it has potential to accumulate in the vital body organs such as liver and kidney. High level of Cd in food is responsible for the abnormal function of kidney, liver and cardiovascular system (WHO, 1996).

Mercury naturally occurs as a liquid metal at favorable pressure and temperature. In the form of ionic salts it occurs in two forms (a) Mercury (I) and (b) Mercury (II). Mercury (I) or mercurous salts are less prevalent in the nature than mercuric salts or Mercury (II). Organometallic compounds of mercury have many industrial and agricultural

applications. The elevated level of mercury can cause numerous severe problems concerning health, that may comprises of nervous disorders as damage to brain, nervous system and renal damage and may cause cardiovascular diseases. The pregnant women and their babies are at highest risk as it affects the fetus of the developing brain.

Table 2: The Sources of Some Important Heavy Metals and their Health Implications

Heavy Metal	Source	Acute Effect	Chronic Effect
- Lead absorption is through GI tract and elimination through kidneys	Food additives and lipstick	Weakness of proximal muscle, bone pain, multiple non-healing fractures and osteoporosis	Kidney damage, loss of balance, loss of appetite, liver dysfunction and weakness
- Arsenic toxic metal that is incorporated into nails and hair - Arsenic has no beneficial role in the human body	Herbicides, insecticides contaminated food and water	Nausea, headache, abdominal pain and diarrhea.	Pigmentation and irritation of skin, Polyneuritis and Blackfoot disease
- Cadmium intake is through contaminated food - High cadmium contents of hair reflects past increased intake of the metal or smoke exposure	Ingestion of highly soluble cadmium compounds or Inhalation of fine dust and fumes	Fever with many other complications such as pneumonitis, edema and death	Congenital abnormalities, dried scaly skin, loss of sense of smell, hair loss and pregnancy toxemia
- Chromium is required for proper function of insulin - Some chromium compounds can be toxic and carcinogenic	Coal and crude oil Cement manufacturing, combustion, copying machine toner and leather tanning	Dyspnea, coughing, Vomiting, skin burns, wheezing, , ulcers, nausea and diarrhea	Bronchitis, soreness Soreness, asthma, nasal itching and Pneumonia
- Mercury is toxic in its elemental, organic and inorganic forms	Anti-septic, anti-mildew agents and battery manufacturing	Dysarthria, ataxia and constricted visual fields	Neurological signs and symptoms

Vegetable Washing for Mitigating Heavy Metals

Traditional practices of vegetables washing to remove dirt and debris before consumption has been assumed to reduce the level of heavy metals and pesticide residues. Several washing mediums such as solutions of chlorine, salts, ozonated water and strong acid have been proven very effective in the mitigation of agro-contaminants residues during commercial vegetable processing. Two to three times washing of vegetables with clean water significantly reduces the heavy metals contents. With washing, greater reduction was for lead and cadmium i.e. 75–100% than those for copper and zinc i.e. 27– 55% (Sing *et al.*, 2004).

Since the dietary intake of contaminated food contribute significantly in the buildup of heavy metals in the body and their detrimental impact becomes apparent only after several years of exposure. Keeping in view persistent nature and cumulative behavior as well potential toxic effects of heavy metals as a result of consumption of vegetables, the present study was designed to assess the status of heavy metals in unwashed vegetables (Cauliflower, spinach, okra and brinjal), and to estimate the technical efficiency of chemical techniques for mitigation of heavy metals in these vegetables.

MATERIALS AND METHODS

Vegetable Sample Procurement

Six samples of each vegetable (Cauliflower, spinach, okra and brinjal) were obtained from supervised, wastewater irrigated field of University of Agriculture, Faisalabad. One kilogram sample of each vegetable was obtained at the point of optimum maturity. The samples were immediately transferred to the laboratory of National Institute of Food Science and Technology, for subsequent analysis of samples.

Sample Preparation

The collected vegetable samples of vegetables were reduced to appropriate size by using knife to facilitate the analysis. The study was designed with five washing treatments by using tap water and following concentrations of chemical solutions in water along with one unwashed sample.

Table 3: Different Washing Treatments

Chemical/ Reagent	Concentration	Treatment
A. Unwashed	-	T ₀
B. Tap water	-	T ₁
Chemical Solution		
C. Acetic acid	5%	T ₂
	10%	T ₃
D. Sodium chloride	5%	T ₄
	10%	T ₅

After solution preparation vegetable sample were soaked in solution for ten minutes for washing purpose. The washed vegetables were blended by using commercial blender to make homogeneous paste of pulp and juice. The vegetable samples (three replicates of each washing treatment) were kept in hot air oven at 70-80 °C till complete dryness. The dried vegetable samples of different parts of vegetables were ground into a fine powder (80 mesh) using a commercial blender and stored in polyethylene bags for analysis.

Sample Digestion

The powdered vegetable sample (0.5-1.0 g) was taken into a Pyrex beaker; 10 mL of concentrated HNO₃ was added to it and kept overnight without heating. It was then heated on a hotplate and after evaporation near to dryness, the sample was cooled and 5 mL HClO₄ was added and heated again. After digestion was complete, the sample was filtered into a clean volumetric flask and diluted to 50 mL with double de-ionized water (Nwajei, 2009).

Preparation of Standards

The standard solutions (2, 4, 6, 8, 10, 15, 20 and 25 ppm) of all the metals (Cd, Cr, As, Pb and Hg) were prepared from the stock standard solutions containing 1000 ppm in distilled water.

Determination of Metals

The heavy metals Cd, Cr, As, Pb and Hg in unwashed, tape water, chemically washed, dried and digested vegetable samples was determined by using Varian GTA 120 AA 240 Graphite Atomic Absorption Spectrophotometer (AOAC, 2006). The data obtained was analyzed and represented using standard statistical procedures i.e. completely randomized design (CRD) as described by Steel *et al.* (1997).

RESULTS

The concentrations of cadmium, chromium, arsenic, lead and mercury were analyzed in six samples of each vegetable (one unwashed and five washed) collected from supervised field at the stage of optimum maturity. The results of undertaken studies are presented and discussed in this section. The results for cadmium, chromium, arsenic, lead and mercury concentration in different vegetables were highly significant as shown in Table 1,2,3,4 and 5 respectively.. The concentration of heavy metals in unwashed vegetable samples were higher than samples subjected to different washing treatments and all treatments exceeded the MRL value for cadmium (0.05 mg kg⁻¹), chromium (1.30 mg kg⁻¹), arsenic (0.1 mg kg⁻¹), lead (0.3 mg kg⁻¹) and mercury (0.03 mg kg⁻¹) set by FAO. Different washing techniques resulted in varying

degree of cadmium reduction in vegetables (Table 1). Out of all the washing treatments, maximum heavy metal reduction was observed in T₃ i.e. 10% acetic acid solution, which reduced the cadmium contents to 23% (Cauliflower), 29% (spinach), 22% (okra) and 26% (brinjal) as compared to unwashed samples. Contrarily minimum reduction was observed in T₂ i.e. tap water which resulted in only 4-6% reduction in cadmium concentration in different vegetables. The reduction pattern of chromium by T₃ i.e. 10% acetic acid was 20, 23, 19 and 21% in cauliflower, spinach, okra and brinjal respectively (Table 2). Similarly unwashed samples of all vegetables were found to be heavily contaminated with arsenic, lead and mercury. Tap water reduce the arsenic, lead and mercury contents by 5, 7 and 7% (Cauliflower), 6, 7 and 9% (Spinach), 4, 6 and 8% (Okra), 5, 5 and 6% (Brinjal) respectively. Again T₃ i.e. 10% acetic acid was found to be more effective in all the vegetables to reduce arsenic, lead and mercury contents as shown in the Table 3, 4 and 5.

Table 1: Cadmium Contents in Different Vegetables (mg/Kg)

Treatment	Vegetables							
	Cauliflower		Spinach		Okra		Brinjal	
	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R
T ₀	0.0641±0.001 ^a	-	0.0714±0.004 ^a	-	0.0571±0.011 ^a	-	0.0593±0.013 ^a	-
T ₁	0.0615±0.004 ^b	4	0.0678±0.009 ^b	5	0.0536±0.005 ^b	6	0.0563±0.033 ^b	5
T ₂	0.0519±0.001 ^e	19	0.0578±0.003 ^e	19	0.0491±0.022 ^d	14	0.0515±0.052 ^c	13
T ₃	0.0490±0.008 ^f	23	0.0506±0.011 ^f	29	0.0445±0.019 ^e	22	0.0438±0.031 ^e	26
T ₄	0.0588±0.006 ^c	8	0.0619±0.007 ^c	12	0.0513±0.003 ^c	10	0.0521±0.011 ^c	12
T ₅	0.0554±0.004 ^d	13	0.0588±0.003 ^d	17	0.0485±0.043 ^d	15	0.4862±0.021 ^d	18

P.R: Percent Reduction

T₀ : Unwashed

T₁: Tap water washed

T₂: Acetic acid 5%

T₃: Acetic acid 10%

T₄: Sodium chloride 5%

T₅: Sodium chloride 10%

Table 2: Chromium Contents in Different Vegetables (mg/Kg)

Treatment	Vegetables							
	Cauliflower		Spinach		Okra		Brinjal	
	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R
T ₀	1.224±0.005 ^a	-	1.907±0.029 ^a	-	1.024±0.001 ^a	-	1.501±0.021 ^a	-
T ₁	1.131±0.009 ^b	6	1.771±0.009 ^b	7	0.983±0.004 ^b	4	1.410±0.011 ^b	6
T ₂	1.055±0.004 ^d	13	1.623±0.004 ^d	14	0.911±0.005 ^d	11	1.275±0.045 ^d	15
T ₃	0.976±0.041 ^f	20	1.461±0.010 ^e	23	0.829±0.019 ^e	19	1.185±0.029 ^e	21
T ₄	1.109±0.005 ^c	9	1.719±0.006 ^c	10	0.952±0.004 ^c	7	1.335±0.007 ^c	11
T ₅	1.039±0.008 ^e	15	1.627±0.007 ^d	14	0.901±0.011 ^d	12	1.260±0.001 ^d	16

Table 3: Arsenic Contents in Different Vegetables (mg/kg)

Treatment	Vegetables							
	Cauliflower		Spinach		Okra		Brinjal	
	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R
T ₀	0.1231±0.008 ^a	-	0.1310±0.012 ^a	-	0.1101±0.022 ^a	-	0.1333±0.003 ^a	-
T ₁	0.1178±0.009 ^b	5	0.1224±0.002 ^b	6	0.1056±0.001 ^b	4	0.1266±0.033 ^b	5
T ₂	0.1062±0.041 ^d	13	0.1154±0.020 ^c	12	0.1001±0.013 ^c	9	0.1199±0.042 ^d	10
T ₃	0.1010±0.009 ^e	18	0.1061±0.034 ^d	19	0.0946±0.045 ^d	14	0.1106±0.051 ^e	17
T ₄	0.1124±0.003 ^c	8	0.1214±0.006 ^b	7	0.1045±0.003 ^b	5	0.1226±0.011 ^c	8
T ₅	0.1060±0.024 ^d	12	0.1156±0.024 ^c	11	0.1012±0.044 ^c	8	0.1213±0.032 ^c	9

Table 4: Lead Contents in Different Vegetables (mg/kg)

Treatment	Vegetables							
	Cauliflower		Spinach		Okra		Brinjal	
	Mean±S.D (Mg/Kg)	P.R	Mean±S.D (Mg/Kg)	P.R	Mean±S.D (Mg/Kg)	P.R	Mean±S.D (Mg/Kg)	P.R
T ₀	0.5722±0.008 ^a	-	0.8783±0.006 ^a	-	0.4120±0.002 ^a	-	0.6715±0.003 ^a	-
T ₁	0.5311±0.003 ^b	7	0.8221±0.012 ^b	7	0.3872±0.009 ^b	6	0.6379±0.011 ^b	5
T ₂	0.4699±0.002 ^d	18	0.7064±0.009 ^d	19	0.3460±0.005 ^d	16	0.5774±0.009 ^c	14
T ₃	0.4151±0.016 ^c	27	0.5846±0.005 ^f	33	0.2842±0.011 ^f	31	0.4767±0.022 ^e	29
T ₄	0.4927±0.008 ^c	14	0.7330±0.016 ^c	16	0.3584±0.013 ^c	13	0.5707±0.013 ^c	15
T ₅	0.4784±0.009 ^d	17	0.6910±0.009 ^c	21	0.3337±0.007 ^e	19	0.5237±0.005 ^d	22

Table 5: Mercury Contents in Different Vegetables (Mg/Kg)

Treatment	Vegetables							
	Cauliflower		Spinach		Okra		Brinjal	
	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R	Mean±S.D (mg/kg)	P.R
T ₀	0.0312±0.005 ^a	-	0.0711±0.006 ^a	-	0.0612±0.008 ^a	-	0.0510±0.012 ^a	-
T ₁	0.0285±0.005 ^b	7	0.0637±0.004 ^b	9	0.0563±0.014 ^b	8	0.0479±0.022 ^b	6
T ₂	0.0265±0.006 ^d	15	0.0589±0.002 ^d	17	0.0514±0.009 ^d	16	0.0418±0.002 ^d	18
T ₃	0.0236±0.004 ^f	24	0.0515±0.005 ^f	27	0.0471±0.001 ^f	23	0.0382±0.009 ^f	25
T ₄	0.0276±0.001 ^c	12	0.0620±0.003 ^c	12	0.0532±0.003 ^c	13	0.0433±0.005 ^c	15
T ₅	0.0256±0.004 ^e	18	0.0578±0.009 ^c	17	0.0496±0.044 ^e	19	0.0402±0.011 ^e	21

DISCUSSIONS

Vegetables play key role in maintaining the nutritional status of humans. The dominant elements in vegetables are calcium, potassium, iron and sodium etc. which provide alkalizing effects, neutralizing the acidity produced by other foods, especially those of animal origin. Safe food production is an important aspect of food quality assurance as well as human health. Heavy metals in vegetables accumulate through uptake from soil, water and atmosphere. The intake of heavy metals through diet leads to several disorders such as kidney damage, nervous disorder, bone disease and tubular growth. Vegetables uptake the metals from contaminated soil by absorbing them as well as from deposits on the vegetable parts exposed to the air from polluted environments. In the present study four vegetables i.e. cauliflower, spinach, okra and brinjal were grown and samples were obtained at the stage of optimum maturity to assess the concentration of heavy metals in unwashed, tap water and chemically washed vegetables.

Washing as a practice is very common and effective in most households as easily available plain water and readily available chemical solutions can be used for vegetable washing in house hold kitchen (Krol *et al.*, 2000). There was about 75 to 100 percent reduction in lead and cadmium and about 27 to 55 percent reduction in copper and zinc contents when the vegetable samples were washed two to three times with clean tap water (Sing *et al.*, 2006).

In the present study, difference in the heavy metal reduction potential of different treatments was due to difference in concentration and type of the chemical agent used for the washing of vegetable. All vegetable samples were found to be heavily contaminated with different metals. The excessive intake of toxic metals can cause severe complications in humans and animals. Consumption of fruits and vegetables loaded with heavy metals such as Cd, Pb or even Cu and Zn are reported to cause cancer. There are already published works related to the incidence of gastrointestinal cancer (Turkdogan *et al.*, 2002) and cancer of the pancreas, urinary bladder or prostate (Waalkes and Rehm, 1994). Lead, cadmium and chromium were found to be accumulated in the shoot and roots of plants at low, medium or high levels (Verma and Dubey, 2003; Yang *et al.*, 2003; Chandra and Kulshrestha, 2004; Adeyeye, 2005).

Leafy vegetables uptake much higher levels of toxic metals as compare to other vegetables because of their more exposure to environmental pollution. Sharma *et al.* (2009) also reported that leafy vegetables were more contaminated with cadmium ($0.09\mu\text{gg}^{-1}$) and the minimum concentration ($0.002\mu\text{gg}^{-1}$) was in cucurbit vegetables (Indian squash). Results also showed that maximum concentration ($0.15\mu\text{gg}^{-1}$) of lead was found in leafy vegetables (coriander) and the minimum concentration ($0.001\mu\text{gg}^{-1}$) in root/ tuberous vegetables (sugar beet). Similar results were observed by Perveen *et al.* (2011) to estimate the concentration of cadmium in unwashed vegetables. The cadmium reduction pattern in this study was parallel to the investigation that was carried out by Singh *et al.* (2006) to estimate the heavy metal load of unwashed and washed vegetables in peri-urban, Delhi. In addition, Suruchi *et al.* (2011) also reported the same reduction pattern of cadmium in spinach, okra and brinjal, exposed to different degrees of pollution in Agra, by washing with tap water. For chromium concentration, results of the present study were supported by Abbas *et al.* (2010) to estimate the concentration of chromium in vegetables and chromium reduction by washing was the same as described by the Nadine *et al.* (2009) in vegetables grown and sold in selected areas in Lebanon.

The arsenic is a toxic element and the humans may get exposure to arsenic through drinking water obtained by wells bored into arsenic contaminated areas or through contaminated water by agro-chemical waste or industrial effluents. Nadine *et al.* (2009) also reported the arsenic reduction by washing of vegetable with tap water but that was slightly high than in present study.

The level of lead is higher than the recommended ($0.2\mu\text{gg}^{-1}$) in humans, cause bones, pancreases, gum, liver, nervous system, teeth and blood diseases. Lead toxicity results in reduction of hemoglobin synthesis, joints pain, kidney damage, defects in reproductive, cardiovascular and nervous systems (Ogwuegbu and Muhanga, 2005). The results of the present study regarding the lead concentration in unwashed vegetables are the same as described by Naser *et al.* (2009) to estimate concentration of different heavy metals in vegetables. Lead reduction by tap water washing follows the same pattern as reported by the Singh *et al.* (2006) to estimate the heavy metal load of unwashed and washed vegetables in peri-urban, Delhi. In past, higher level ($0.02\mu\text{gg}^{-1}$) of mercury was found in fenugreek/methi i.e. leafy vegetables and the lower contents ($0.001\mu\text{gg}^{-1}$) were found in root/tuberous vegetables and fruity vegetables (okra) was found to be heavily contaminated with arsenic ($0.083\mu\text{gg}^{-1}$) whereas the minimum level ($0.014\mu\text{gg}^{-1}$) was detected in cauliflower (Nergus *et al.*, 2005). Results of the present study for lead reduction by tap water washing differ from the findings of the Suruchi *et al.* (2011) to assess the heavy metals concentration in washed and unwashed vegetables exposed to different degrees of pollution in Agra, India.

CONCLUSIONS

The heavy metal residues were found in cauliflower, spinach, okra and brinjal at varying concentrations. Wastewater is the single largest contributor in the heavy metals accumulation in soil as well as in vegetables. In all vegetables, mercury, lead, cadmium, arsenic and chromium residue exceeded the prescribed MRLs in vegetables by FAO/WHO. Spinach show higher accumulation of different metals because of its large surface area as compared to other vegetables. The washing of vegetables with tap water and household chemical solutions not only remove the dirt and dust particles but also reduced the heavy metals significantly. Difference between washed and unwashed vegetables with regard to heavy metal concentrations suggests that heavy metals reaches on the vegetables by aerial deposition and adhere to them. Washing treatments mechanically remove the heavy metals deposited on the surface of the vegetables. On the basis of present study, it is strongly recommended that vegetables must not be grown with sewerage water and vegetables must be washed carefully before cooking to decrease the intake of heavy metals.

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